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Temperature-Dependent Reflectivity of Silicon Carbide

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TEMPERATURE-DEPENDENT REFLECTIVITY OF SILICON CARBIDE

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SUMMARY

The spectral reflectivity of a commercial silicon carbide (SiC) ceramic surface was measured at wavelengths from 2.5 to 14.5 μm and at temperatures ranging from 358 to 520 K using a NASA-developed multiwavelength pyrometer. The SiC surface reflectivity was low at the short wavelengths, decreasing to almost zero at 10 μm, then increasing rapidly to a maximum at approximately 12.5 μm, and decreasing gradually thereafter. The reflectivity maximum increased in magnitude with increasing surface temperature. The wavelength and temperature dependence can be explained in terms of the classical dispersion theory of crystals and the Lorentz electron theory. Electronic transitions between the donor state and the conduction band states were responsible for the dispersion. The concentration of the donor state in SiC was determined to be approximately $4x10^{18}$ and its ionization energy was determined to be approximately 71 meV.

INTRODUCTION

Ceramics are expected to be used in the design of the next generation of advanced propulsion systems. Accurate surface temperature measurements of these materials in engine environments is urgently needed for their research and development.

Pyrometry is a remote temperature measurement method. It measures temperature by measuring the thermal radiation emitted by a surface using Planck's law of radiation. Traditional pyrometers are either the one- or the two-color variety. The one-color pyrometer measures the radiation at one narrow wavelength band. The surface emissivity must be known for accurate temperature results. The two-color pyrometer measures the radiation at two narrow wavelength bands and does not require knowing the surface emissivity; instead, the surface emissivities at the two bands are assumed to be equal.

Both types of pyrometers are susceptible to interference from extraneous radiation, which is of great concern when highly reflective surfaces are involved (ref. 1). NASA Lewis Research Center has developed a multiwavelength pyrometer to measure the surface temperatures of ceramic materials such as silicon carbide (SiC). This pyrometer has been shown to be applicable even in the presence of interfering radiation (refs. 2 and 4).

After developing this pyrometer, we have used it to measure the surface temperature of a silicon carbide ceramic sample and to obtain simultaneously its reflectivity and emissivity. The measured reflectivity can be compared to a theoretical value using the classical dispersion theory for crystals.

EXPERIMENTAL METHOD

The NASA-developed multiwavelength pyrometer (ref. 3) has been applied to measure the temperature of silicon carbide and to investigate the temperature dependence of its reflectivity and emissivity. The experimental arrangement was the same that was used previously and is shown in figure 1. In general, the location of the spectrometer and the auxiliary radiation source is specified by angular coordinates (β,Θ) where β is the angle measured from the surface normal to the surface, and Θ is the angle above the plane of the figure. In these experiments, the spectral radiometer was positioned to view the surface at an angle of about 30° measured from the surface normal. The auxiliary radiation source was located at approximately the same angle on the opposite side of the surface normal. Thus, the directional reflectivity and emissivity measured correspond to β=30° and Θ=0°.

The SiC sample used in the study is described in reference 3. Its dimensions were 25 by 50 by 3 mm (1 by 2 by 1/8 in.) and it was gray to black in appearance with a nominal surface finish of 10 μm. The sample was positioned at the opening of a blackbody furnace cavity so that is was completely covered. The temperature of the sample is the equilibrium temperature resulting from the blackbody furnace heat flux incident on the back surface of the sample and heat loss from the front surface. For each temperature measurement, two radiation spectra were collected and analyzed (ref. 4) to determine the sample temperature.

Successful temperature measurements were made up to 517 K. Above this temperature, the surface-emitted radiation overwhelmed that from the 22-W auxiliary radiation source. As a result, the fractional increase in radiation was less than the resolution of the spectral radiometer. When this happens, the procedure of obtaining the reflected spectrum by subtraction of two spectra (ref. 3) is no longer feasible. This difficulty will be overcome in a future system design in which the reflected spectrum can be obtained from a single measuring operation by mechanically chopping the auxiliary radiation and using phase-sensitive detection.

Shown in figure 2 are the SiC surface emission spectra of each of the measurements, the Planck curve corresponding to the temperature as measured by the multiwavelength pyrometer, and the fitted curve obtained by modifying this Planck curve using the emissivity values determined during curve fitting. The fitted curve (modified Planck curve) at each temperature agrees well with the measured spectrum. We previously attached a thermocouple to the sample surface and demonstrated that a good fit to the spectrum is an indication of accurate temperature measurement (refs. 3 and 4).

As a result of measuring the SiC surface temperature by curve fitting, the reflectivity and emissivity of the SiC surface (β = 30°, Θ = 0°) at these temperatures were automatically obtained. Their spectral and temperature dependence are shown in figures 3 and 4, respectively.

THEORY AND DISCUSSIONS

The SiC surface was not very reflective in the wavelength region between 2.5 and 10 μm as shown in figure 3. The temperature dependence was also very weak in this region. Above 10 μm, both wavelength and temperature dependence were more pronounced. The reflectivity peak occurred at a wavelength of approximately 12.13 μm and its magnitude increased from 40 percent at the lowest temperature to 70 percent at the higher temperature. An emissivity minimum occurred at about

12.5 μm and decreased with increasing temperature. This minimum was also reported in reference 5. The surface finish of those samples were reported to be rough, resembling that of a diamond wheel cut. The emissivity was measured by comparing the radiation from the specimen surface with that from a cavity at the same temperature.

A broad, unresolved peak at approximately 10 μm was also observed by Atkinson et al. who measured the emissivity of silicon carbide using an emissometer at discrete wavelengths (ref. 6). Because of the difficulties inherent in this measurement procedure, previous emissivity measurements on SiC and other nongray ceramic surfaces were done only at discrete wavelength regions and at wavelengths below 10 μm (unpublished research by W. Atkinson of Pratt and Whitney).

Spitzer et al. (ref. 7) have published reflectivity data on green, alpha (hexagonal) SiC. The results were obtained for optic axis-oriented crystalline samples using polarized radiation. The data were satisfactorily analyzed using the classical dispersion theory of crystals (ref. 8) and the Lorentz electron theory. The classical dispersion theory gives a theoretical reflectivity r, for normal incidence as

$$r = \frac{(n - 1)^2 + k^2}{(n + 1)^2 + k^2} \tag{1}$$

where

$$n^2 = (1/2)\left\{ [\varepsilon^2 + 4(\sigma/\nu)^2]^{1/2} + \varepsilon \right\} \tag{2}$$

$$k^2 = (1/2)\left\{ [\varepsilon^2 + 4(\sigma/\nu)^2]^{1/2} - \varepsilon \right\} \tag{3}$$

$$\varepsilon = \varepsilon_0 + 4\pi\chi \tag{4}$$

$$\chi = \rho \frac{1 - \nu^2}{(1 - \nu^2)^2 + \gamma^2\nu^2} \tag{5}$$

$$\frac{\sigma}{\nu} = 2\pi\rho \frac{\gamma\nu}{(1 - \nu^2)^2 + \gamma^2\nu^2} \tag{6}$$

characterized by only four quantities, ε_0, ν_0, γ, and ρ, (the high frequency dielectric constant, the resonance frequency, the resonance width, and the resonance strength, respectively) through their relation to the susceptibility χ, the conductivity σ, the index of refraction n, and extinction coefficient k. The quantity ν is the measured frequency divided by ν_0.

Results of applying the classical dispersion theory to analyze the reflectivity data obtained at 517 K are shown in figure 5. By trial and error, the values for the four quantities were determined to be $\varepsilon_0 = 1.6$, $\nu_0 = 2.35\text{x}10^{13}$ Hz, $\rho = 0.045$ and $\gamma = 0.08$ and were used to calculate the reflectivity. By applying the result for normal incidence, we have ignored the angular dependence in the reflectivity. Siegel et al. (ref. 9) calculated the directional emissivity $\varepsilon(\beta)=1-r(\beta)$ of an optically smooth, opaque dielectric and found that the emissivity does not exhibit strong angular dependence for values of β less than 50°. In view of this, the agreement is satisfactory.

The Lorentz theory describes the motion of an electron as an oscillator bound to the equilibrium position in a crystal by Hooke's law forces for which the force constant is κ. The oscillating electron is subject to a periodic electric field of frequency ν as well as to a velocity-dependent damping force with damping constant $2\pi m\gamma$. The theory gives an expression for ρ as

$$\rho = \frac{Ne^2}{4\pi^2 m\nu_0^2} \tag{7}$$

where N is the concentration of ion pairs in the solid, e is the electronic charge, m is the electronic mass, and ν_0, the natural frequency of the oscillator, is given by

$$\nu_0 = \left(\frac{\kappa}{4\pi^2 m}\right)^{1/2} \tag{8}$$

Denoting R_{max} as the maximum reflectivity, Spitzer showed that

$$R_{max} \sim 1 - \frac{\gamma}{\pi\rho} \tag{9}$$

or

$$\begin{aligned}(1 - R_{max})^{-1} &= (\pi/\gamma)\rho \\ &= (\pi/\gamma)\frac{e^2}{4\pi^2 m\nu_0^2}N\end{aligned} \tag{10}$$

The ion pairs in this theory are assumed to originate from thermal excitation of donor (or acceptor) states in silicon carbide. From solid-state physics, the equilibrium concentration of conduction electrons at temperature T, as a result of ionized donors (or acceptors), is given by (ref. 10)

$$N = (n_0 N_d)^{1/2} \exp(-E_d/2kT) \tag{11}$$

with $n_0 = 2(2\pi kmT/h^2)^{3/2}$. Here, N_d is the concentration of donors, E_d is the donor ionization (activation) energy, k is the Boltzmann constant, and h is Planck's constant. Equation (11) holds for $kT << E_d$.

By substituting equation (11) into equation (10) we obtain

$$\begin{aligned}(1 - R_{max})^{-1} &= (\pi/\gamma)\frac{e^2}{4\pi^2 m v_0^2}(2N_d(2\pi km/h^2)^{3/2})^{1/2}\, T^{3/4} \exp(-E_d/2kT) \\ &= A\, T^{3/4} \exp(-E_d/2kT)\end{aligned} \tag{12}$$

At each temperature for which the reflectivity of SiC was measured, the maximum reflectivity was measured (all at wavelength 12.13 µm), the quantity $(1-R_{max})^{-1}$ at the different temperatures was calculated, and then least-squares fitted to equation (12). From curve fitting, A and $E_d/2k$ were determined to be 0.0585 (±0.0014) and 415.77 (±11), respectively. The experimental data and the fitted relation are shown in figure 6. From these results, values of $N_d = (3.7 \pm 0.08) \times 10^{18}$ cm^{-3} and $E_d = 71$ (±1.8) meV were obtained. A value of the order 10^{19} cm^{-3} is reasonable for shallow donors (assumed to be nitrogen) in the noncrystalline SiC that we used. The activation energy of such donors in crystalline SiC is about 90 meV. The value of $E_d = 71$ meV is reasonable because in situations where the concentration is of this magnitude and the nature of the material is noncrystalline, the activation energy is effectively decreased. For the temperatures involved here, the condition $kT << E_d$ is satisfied. For $kT >> E_d$, the result becomes (ref. 5)

$$N \simeq N_d \tag{13}$$

which means that as the temperature increases, the concentration of ion pairs will not increase further, and the reflectivity will not increase beyond 100 percent.

CONCLUSION

The multiwavelength pyrometer developed at NASA Lewis Research Center for measuring nongray surface temperatures was applied to measure the temperature of a silicon carbide surface from 358 to 517 K. The spectral reflectivity and emissivity of silicon carbide at those temperatures and at angles $\beta = 30°$ and $\Theta = 0°$ were also obtained. The emissivity varies rapidly with temperature and wavelength at wavelengths longer than 10 μm.

The reflectivity of SiC obtained during surface temperature measurement using a multiwavelength pyrometer was analyzed using the classical dispersion theory of crystals. The reflectivity maximum at each temperature was fitted to a $T^{3/4} \exp(-E_d/2kT)$ temperature dependence. The curve-fitting-determined constants yielded a donor concentration of 4×10^{18} cm^{-3} and a donor ionization (activation) energy of 71 meV.

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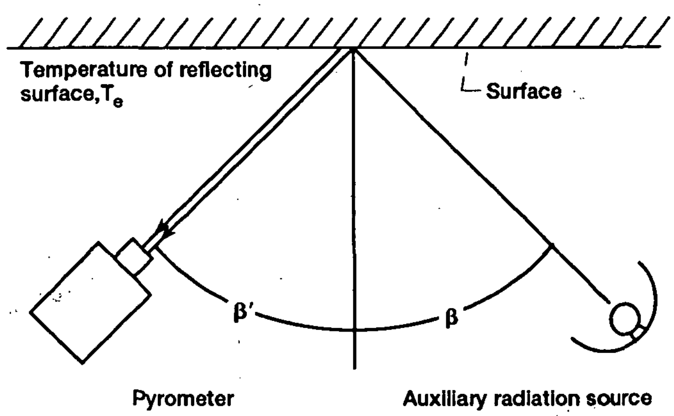

Figure 1.—Experimental arrangement of the pyrometer components. β is an angular direction referred to the surface normal; Θ (not shown) is an angle measured from the plane of the figure.

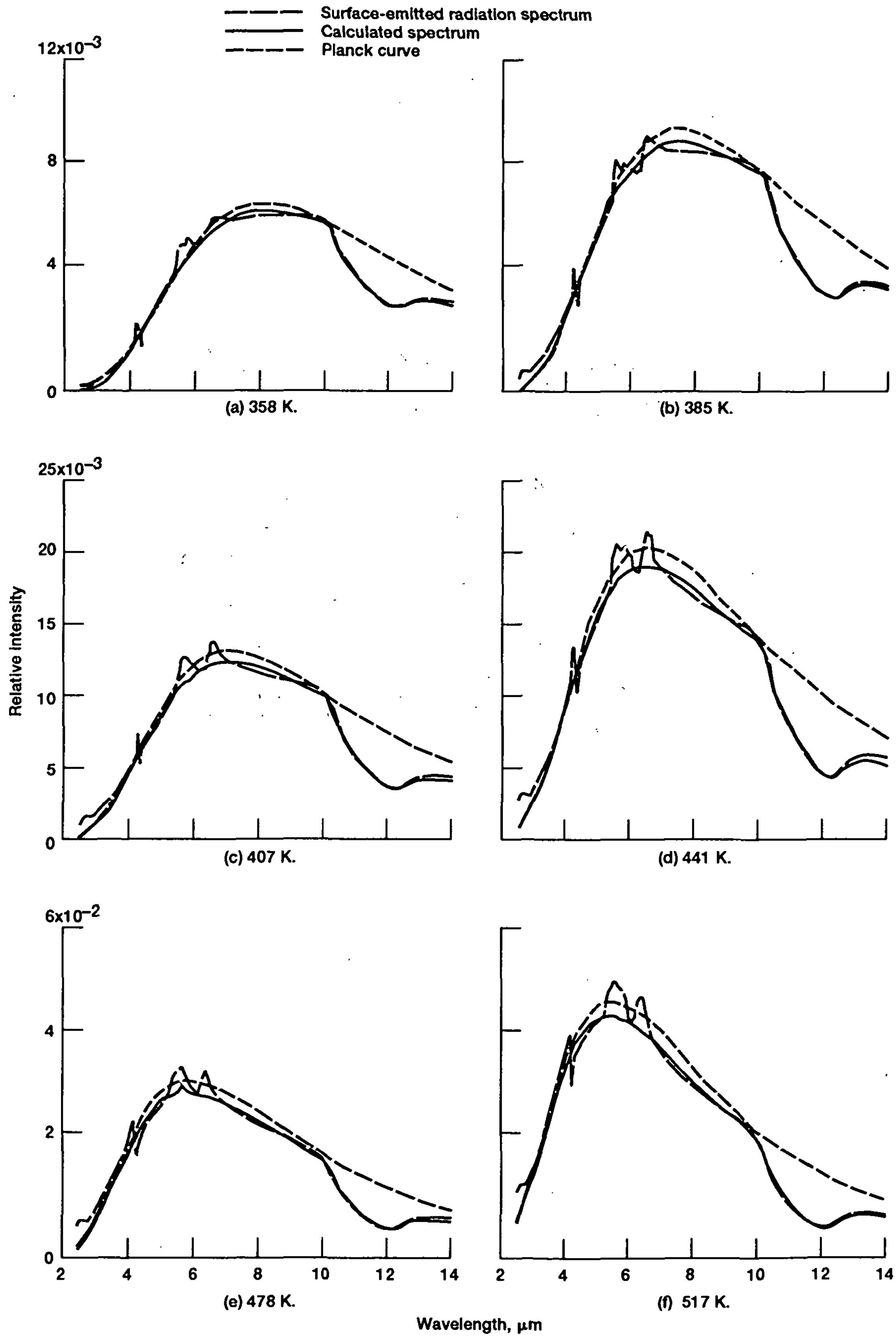

Figure 2.—Surface emission spectrum of SiC compared with calculated spectrum and Planck curve for same temperature.

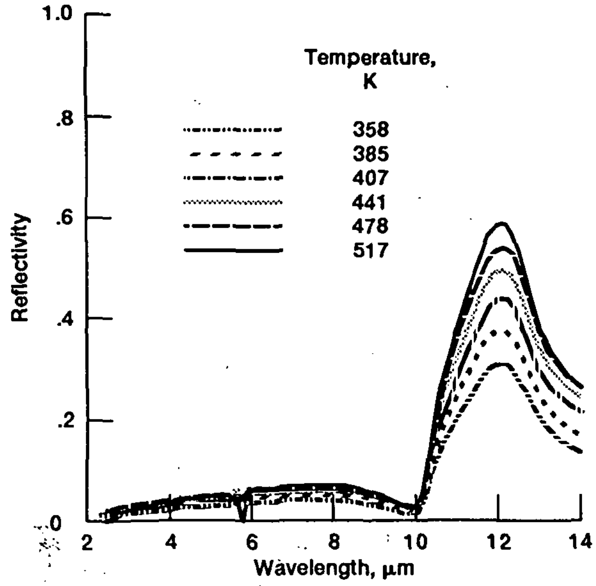

Figure 3.—Spectral reflectivity of SiC at temperatures from 358 to 517 K.

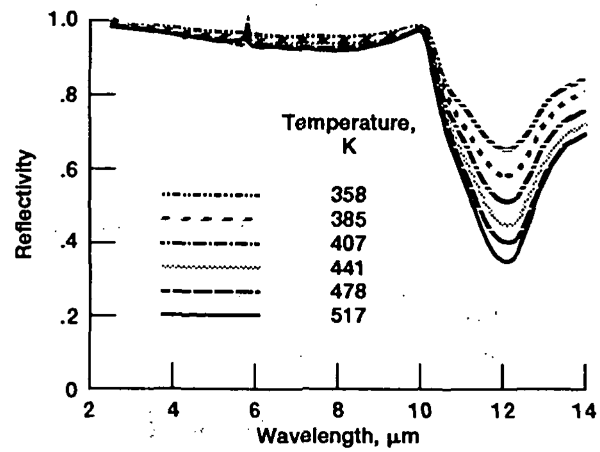

Figure 4.—Spectral emissivity of SiC at temperatures from 358 to 517 K.

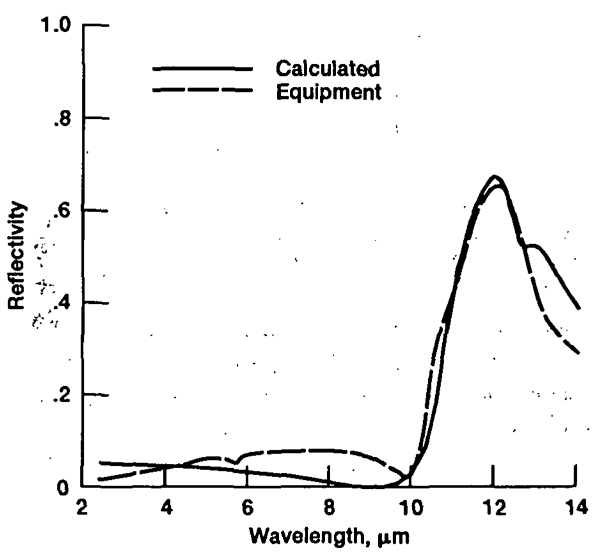

Figure 5.—Experimentally detemined reflectivity compared with calculated reflectivity using the classical dispersion theory with $\nu_0 = 2.35 \times 10^{13}$ Hz, $\rho = 0.045$, $\gamma = 0.08$, and $\epsilon_0 = 1.6$.

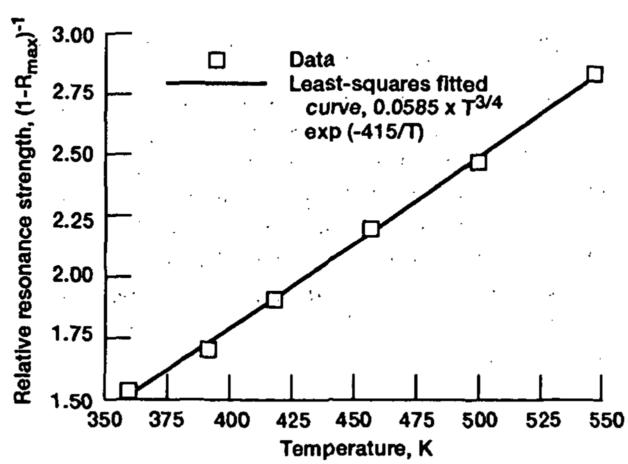

Figure 6.—Relative resonance strength vs temperature.

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